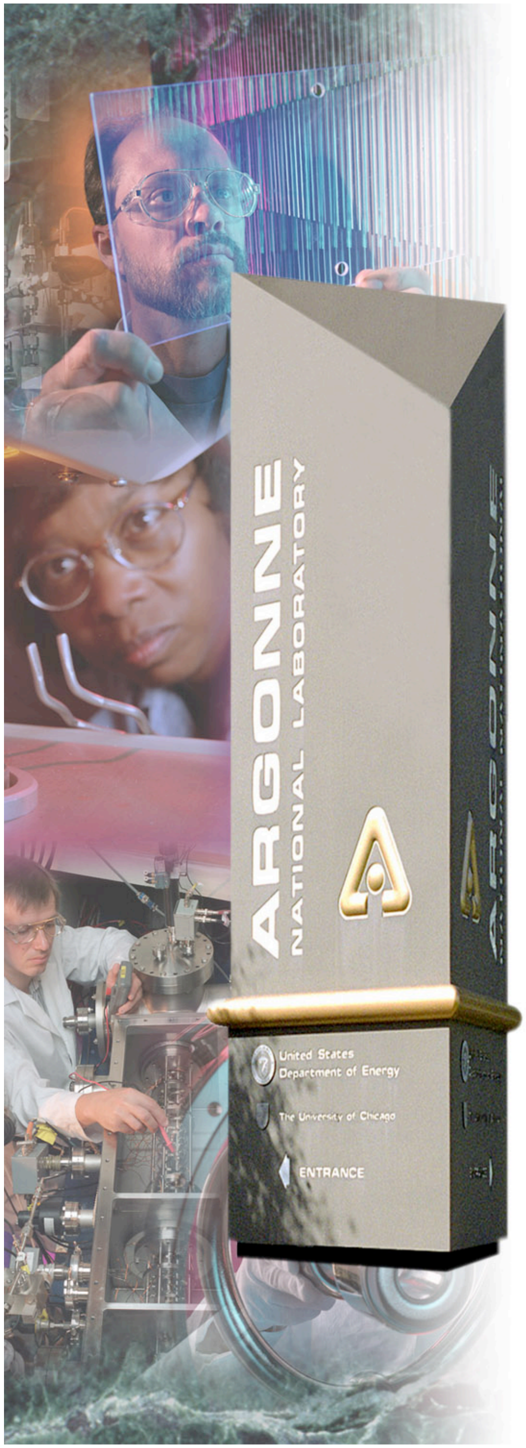


X-raying atoms and molecules in strong optical fields

Linda Young

APS Cross-cut Review

Jan 26, 2005

Argonne National Laboratory

Office of Science
U.S. Department of Energy

A U.S. Department of Energy
Office of Science Laboratory
Operated by The University of Chicago

Collaborators - Sector 7 *(PUP-37 started fall 2004)*

Atomic Physics Group

Bob Dunford, Elliot Kanter, Bertold Krässig,
Steve Southworth, Linda Young
Juana Rudati (APS), Dave Ederer (Tulane)

X-rays

Eric Dufresne, Dohn Arms, Don Walko,
Bernhard Adams (XOR)
Peter Eng (Geo-CARS)

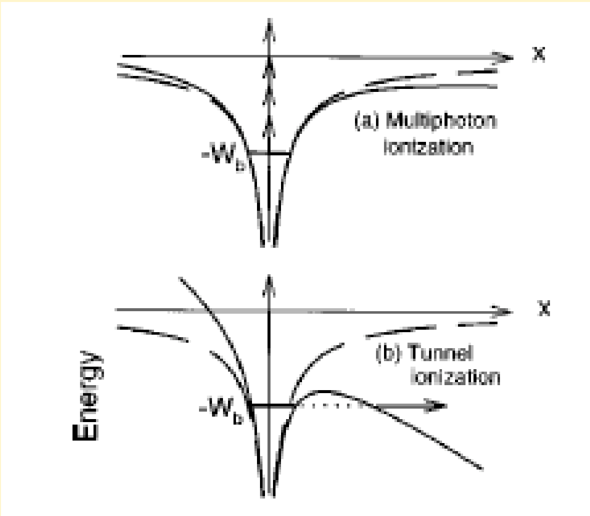
Ultrafast Laser

David Reis, Matt DeCamp (U Michigan)
Rob Crowell, Dave Gosztola (CHM)
Eric Landahl (XOR)

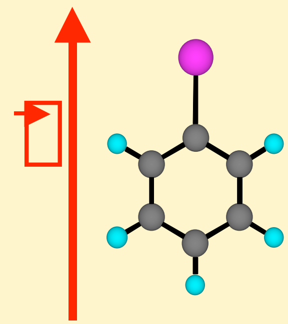
AMO with strong-optical fields

- *Studies of individual atoms and molecules in the gas phase offer simplest rigorous connection with theory*
- *Strong-field effects on inner-shell phenomena never studied*
- *Provide knowledge base for next generation sources*
- *Proposed studies bridge fundamental to applied*

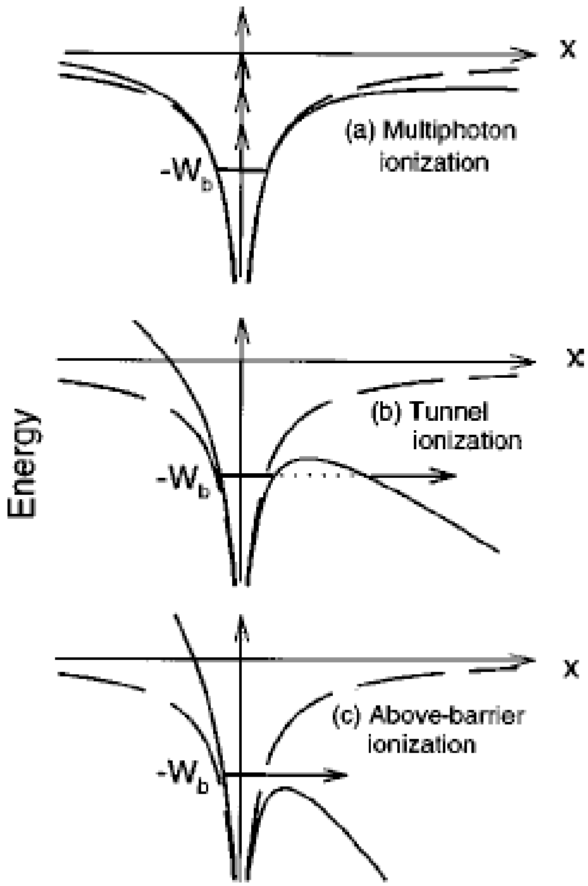
Perturbed x-ray spectra



Aligned molecules



Keldysh picture of atomic response

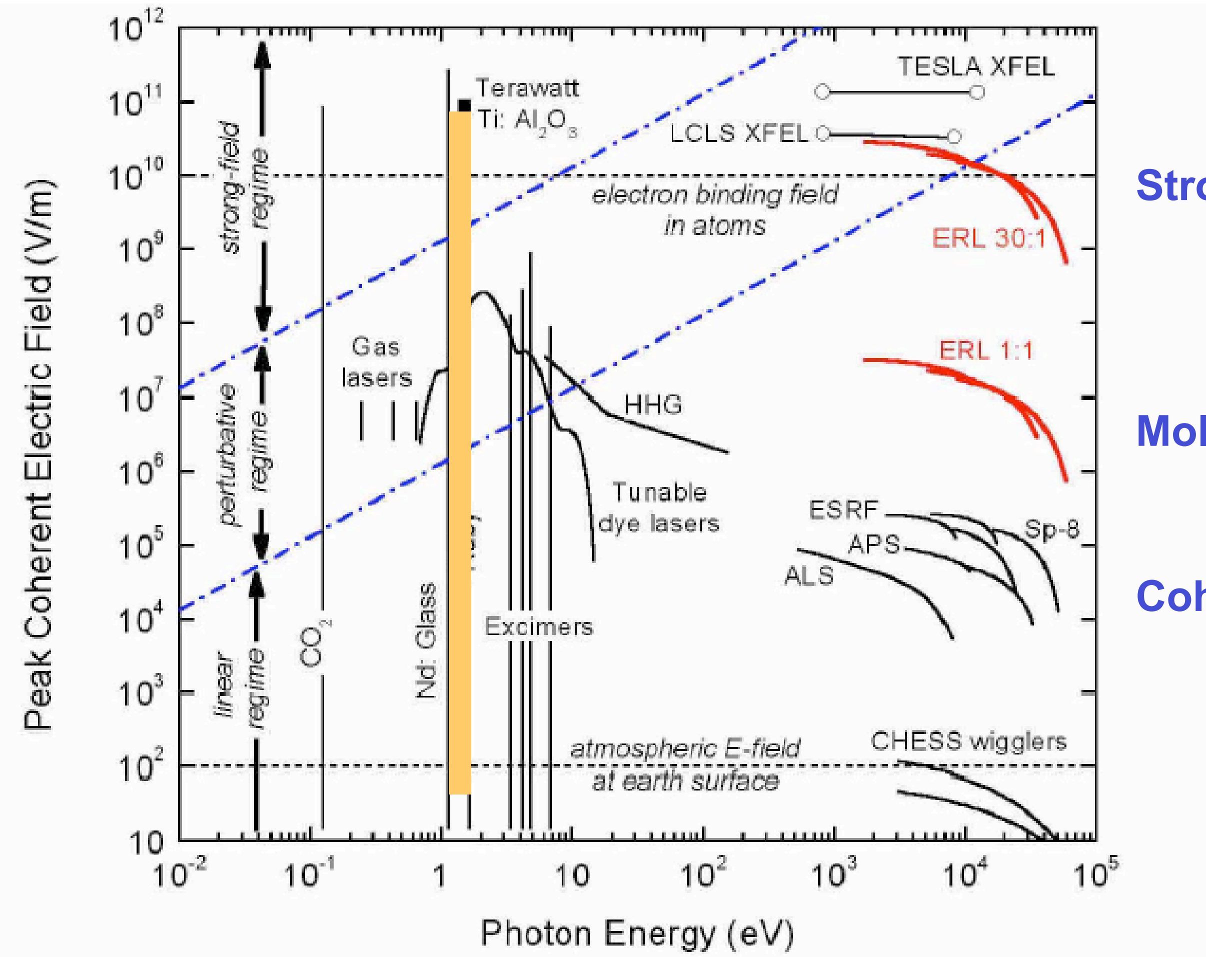


perturbative

$$\gamma \equiv \frac{\text{optical frequency}}{\text{tunnelling frequency}} = (I_p/2U_p)^{1/2}\ \mu\ \ (E\omega)^{-1}$$

"strong-field"

Strong-field landscape



From Q. Shen

Strong-field perturbed spectra

I ~ 10^{15} - 10^{13} W/cm^2

E ~ 9 - 0.9 V/Å

Molecular alignment

I ~10^{13} - 10^{11} W/cm^2

Coherent control

I ~10^9 W/cm^2

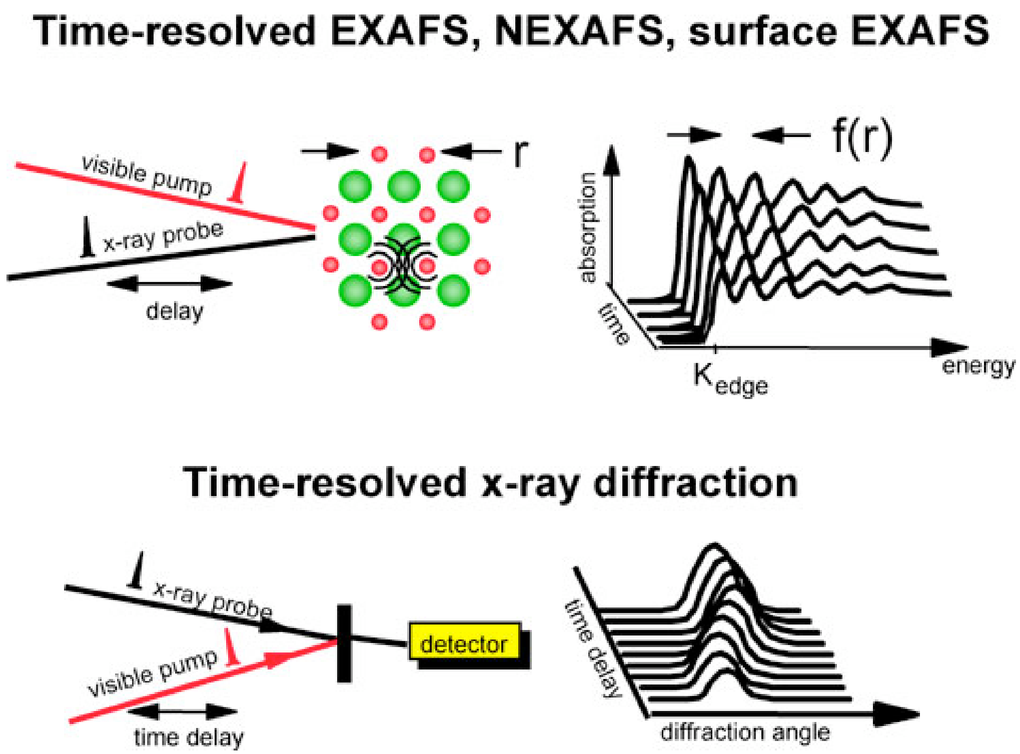
X-ray facilities: toward ultrafast & ultraintense

Single bunch specs

Advanced Photon Source	**10^8 x-rays**	**87 ps**
SPPS Sub-Picosecond Photon Source	**≈10^7 x-rays**	**80 fs**
15 GeV Electrons LCLS 1.5 Å X-Ray Linac Coherent Light Source	**10^{12} x-rays**	**230 fs**

Ultrafast x-ray science

LUX website



Laser-pump/x-ray probe techniques central
Short pulses automatically yield high intensities
1 mJ/100 fs/(0.1 mm)^2 ≈ 10^{14} W/cm^2 ≈ 3 V/Å

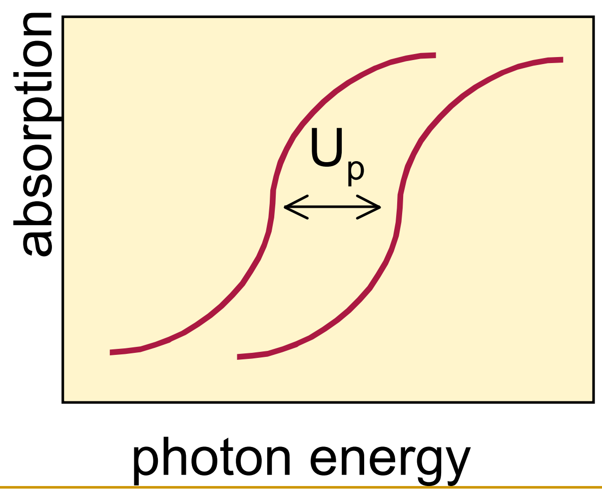
Ultrafast laser/x-ray interactions: isolated atoms

- ***X-ray photoionization is fairly well understood in the weak-field limit***
- ***Understand changes to x-ray processes in presence of strong laser fields***
- ***Theoretical predictions***

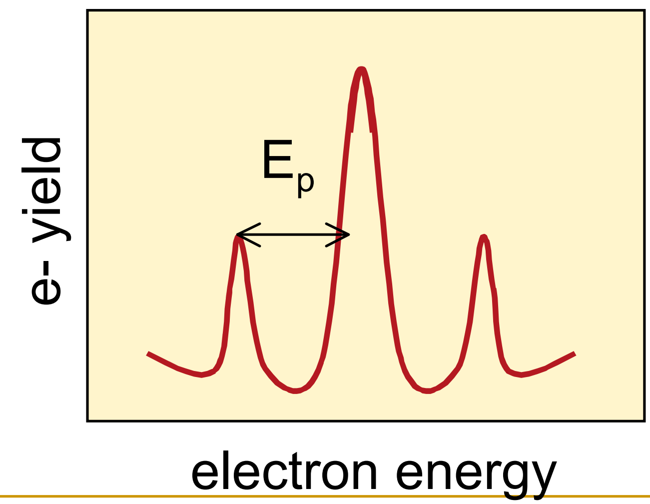
  ponderomotive shift in threshold -> absorption spectrum

  free-free transitions in continuum -> electron spectra

Ponderomotive shift



Electron satellites



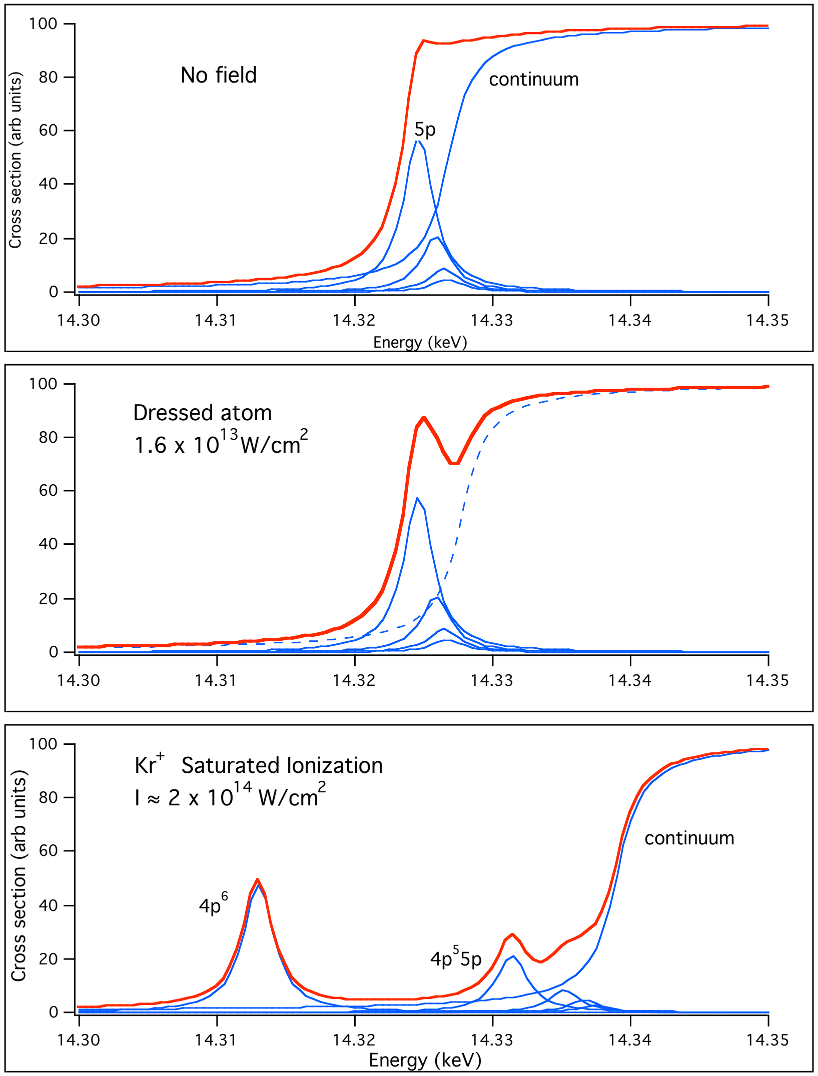
Evolution of Kr 1s-edge structure

- *Naked atom: I=0*
- *Dressed atom: $I<I_{app}$*
- *Ion spectrum: $I>I_{sat}$*

For Kr: 800 nm

$I_{sat} \approx 2 \times 10^{14}$ W/cm^2

$U_p \approx 12$ eV

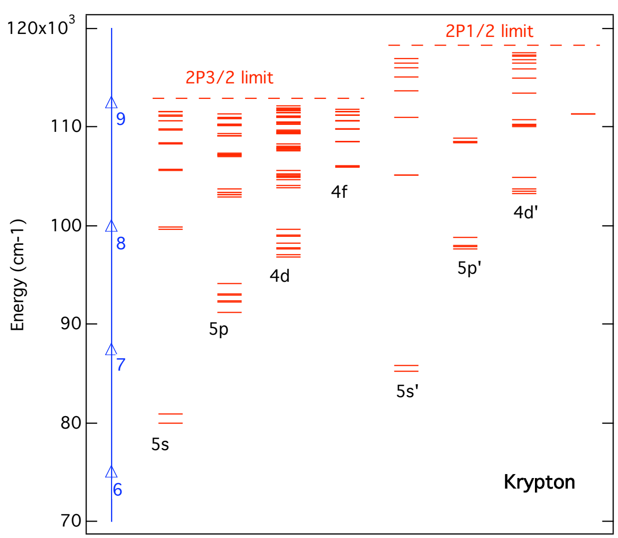


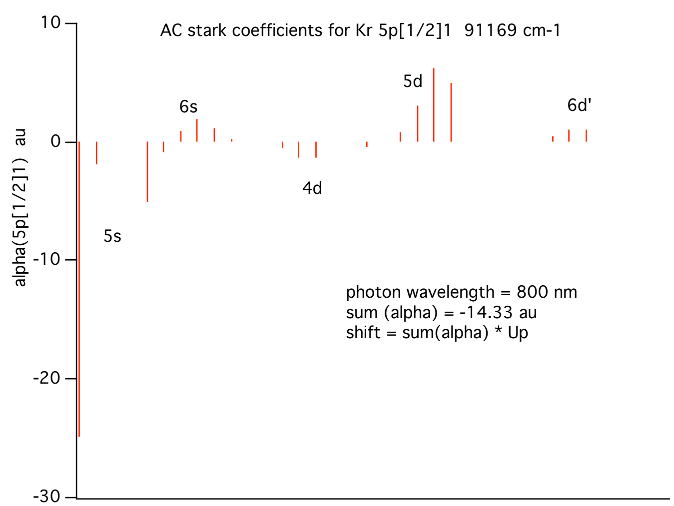
Perturbative AC stark shift for dressed atom

- X-ray absorption probes 1s-np transitions
- Unlike electron spectroscopy which shows all levels
- Shift for 5p levels dominated by 5s-5p interaction

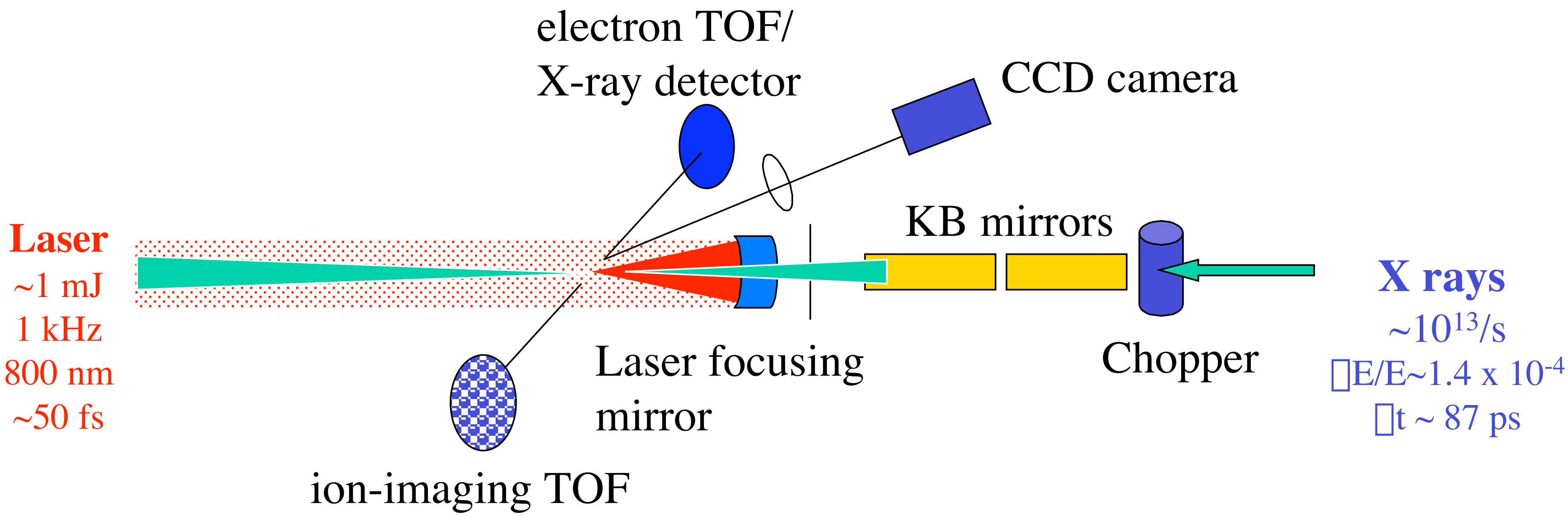
□$W_{5p1/2,gs}$ ~ -14 U_p = -14 eV (for 1.3 x 10^{13} W/cm^2)

$$\Delta E_m = \left(\frac{e\mathbf{E}_0}{2\mu\omega}\right)^2 \sum_n^{\infty} \left[\left| \left\langle n \left| \sum_i^Z \mathbf{x}_i \cdot \hat{\epsilon} \right| m \right\rangle \right|^2 \times \frac{2(E_m - E_n)^3}{(E_m - E_n)^2 - (\hbar\omega)^2} \left(\frac{\mu}{\hbar}\right)^2 \right]$$





Experimental schematic



Laser intensity for 10^{14}W/cm^2 w/1 mJ

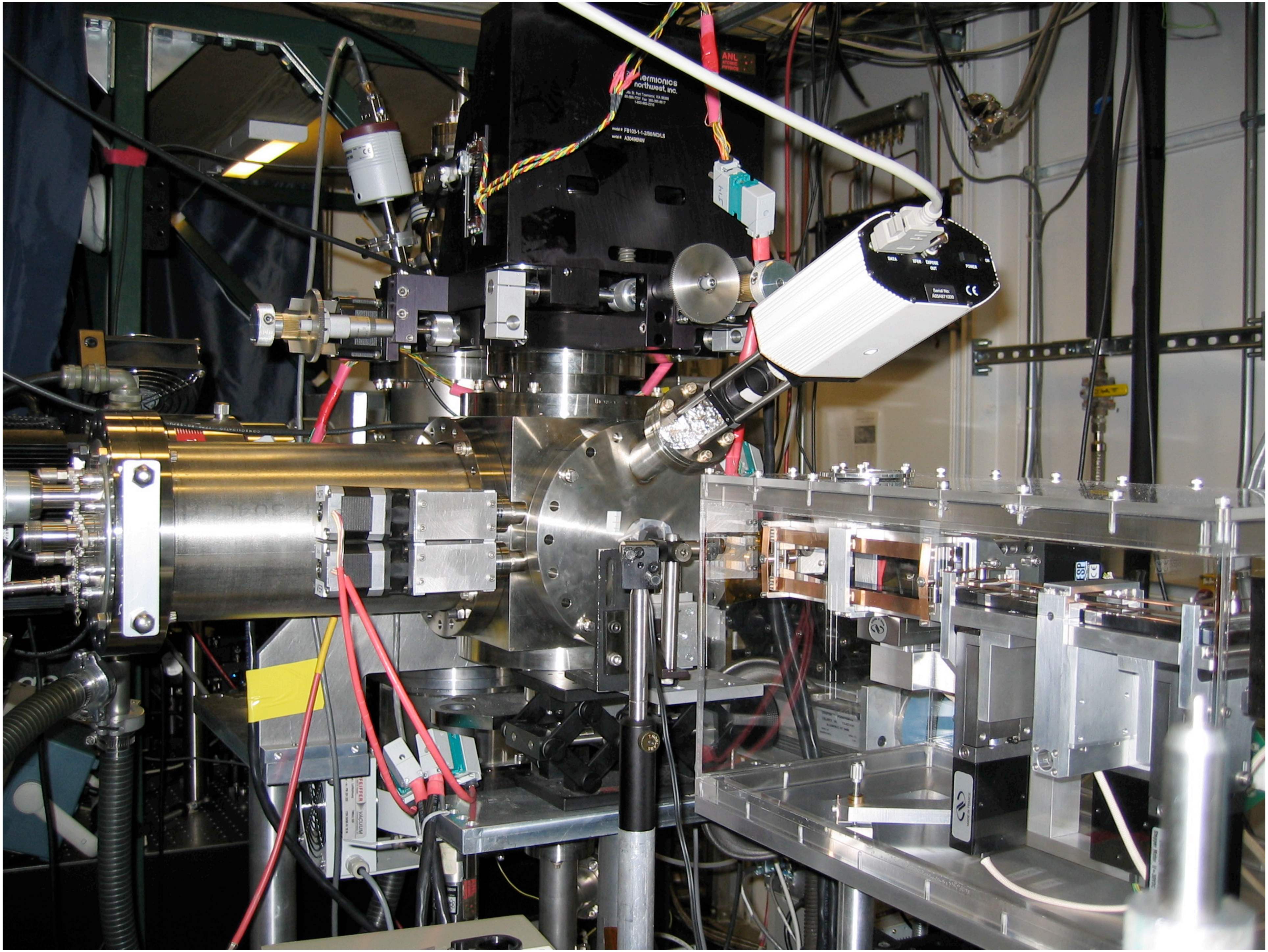
Short pulse (100 fs) focus to ≈100 µm

Long pulse (100 ps) focus to ≈3 µm

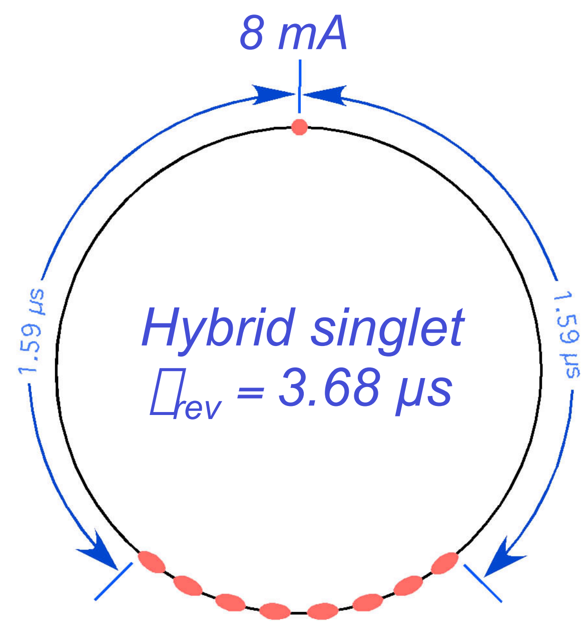
Count rate for gas phase target

10^{13}/cm^3 (vs ~10^{18} in mM solns)

1/3840 x-ray flux coincident

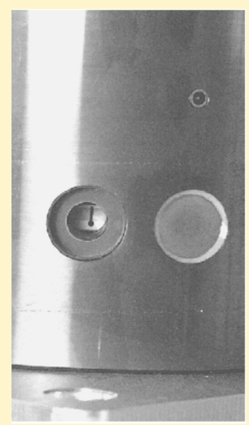

northwest, inc.
ANL

Chopper isolates x-rays coincident with laser



Only 1/3840 of x-ray flux is coincident with laser (overlap with singlet (8/100 total flux) @ 272 kHz)

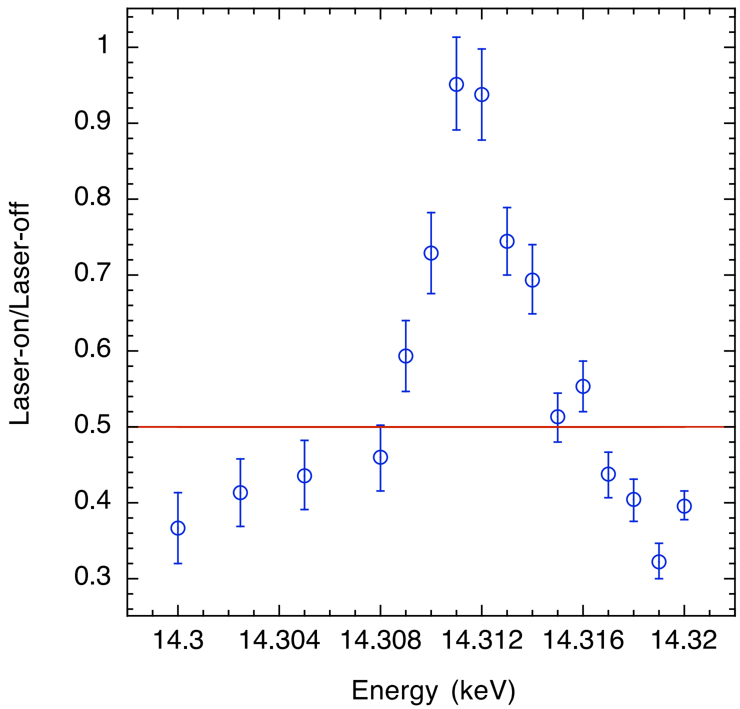
Chopper selects singlet x-ray pulses @ 2.66 kHz
Laser @ 887 Hz : 1 laser-on vs 2 laser-off



0.51 mm slot
50.8 mm diameter
2.45μs open time

First results! Comparison to theory

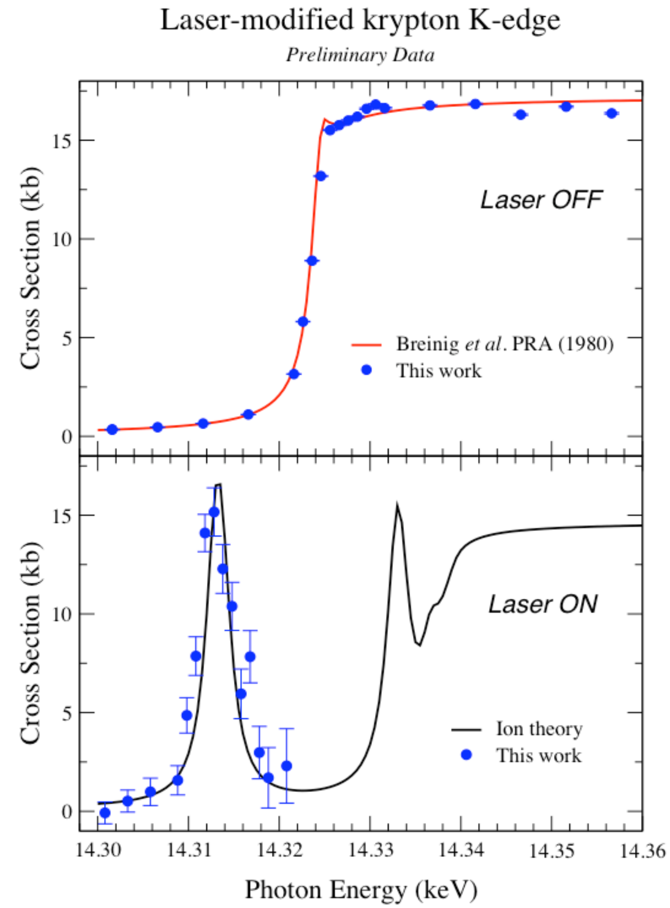
Kα x-ray fluorescence rate



887 Hz laser pulses

2661 Hz x-ray pulses

on/off = 0.5



Neutral Kr

Kr^{1+}

Γ_{1s} = 2.7 eV

Kr^{+} theory: L. Pan and D.R. Beck

Coulomb Explosion of Kr ion assembly

ion production

focal volume ≈30 μm dia × 3 mm long

intensity ≈6 × 10^{14} W/cm^2

atom density ≈10^{13}/cm^3

⇒ 10^7 ions/pulse

electrostatic potentials ≈10 – 100 V

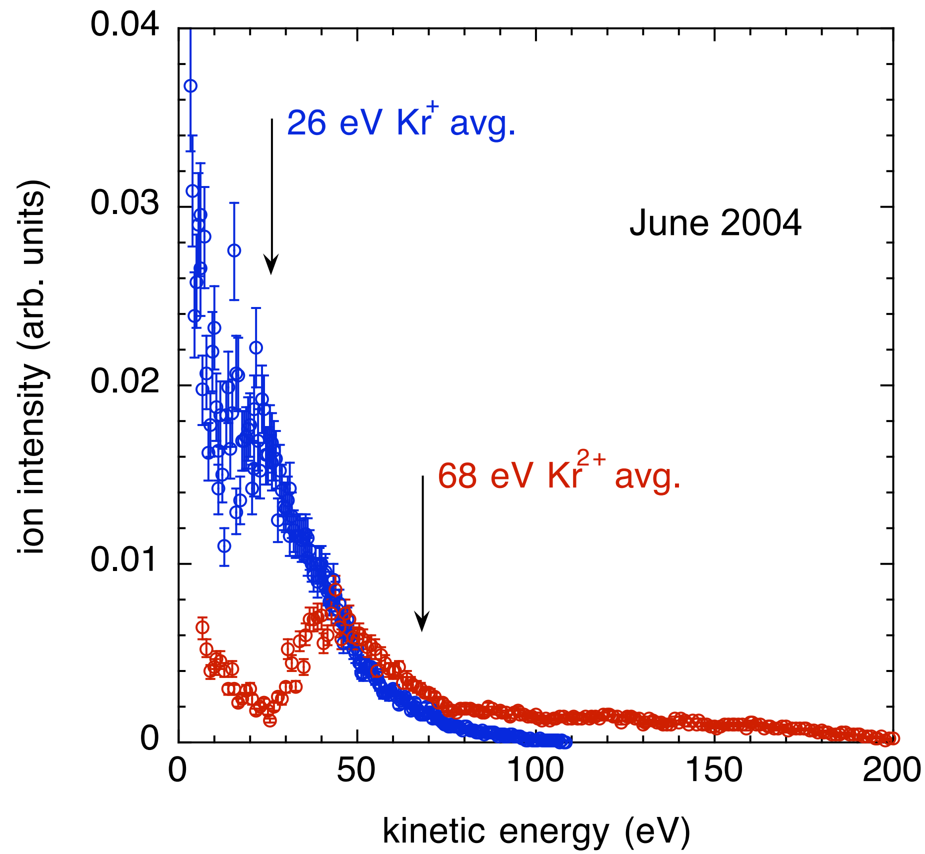
velocities

3.7-eV photoelectron: 1100 μm/ns

26-eV Kr^+: 7.7 μm/ns

68-eV Kr^{2+}: 13 μm/ns

thermal Kr: 0.3 μm/ns



⇒ study expansion dynamics by varying x-ray probe delay

10 µm spatial resolution, 100 ps temporal resolution

Recent Progress - Nov 2004

- *New Ti:sapphire regenerative amplifier (CHM & XFD)*

•2.5 W compressed, 1-5 kHz,
•Diode-pumped
•45 fs FWHM single shot
•27 nm FWHM
(FTL = 35 fs FWHM)

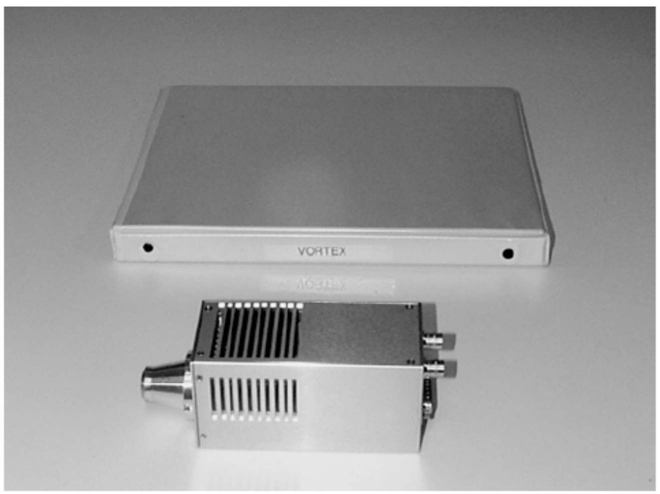
Continuously run Nov 25-Dec 13

- *New Kirkpatrick-Baez mirrors*

- *First active stabilization for x-ray monochromator during scan*
  Before: 10μm/eV beam translation *After: Stable to 1μm*
  Enables 70 eV continuous scan

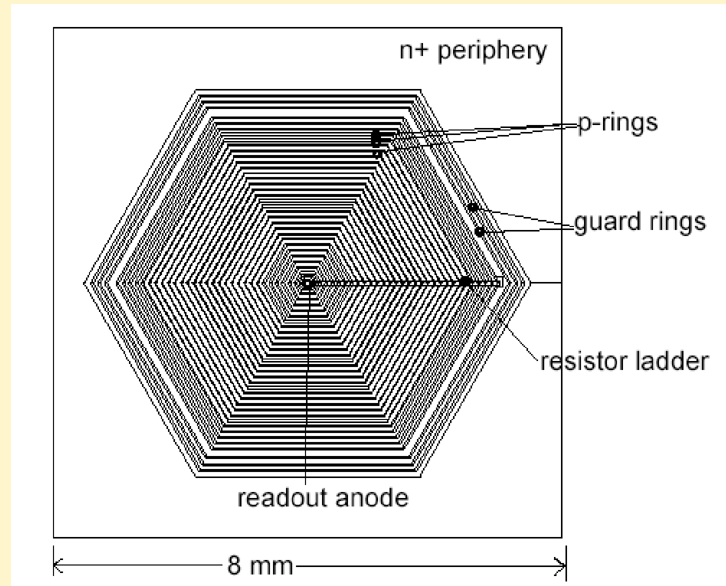
Chopperless operation: photon tagging

- *x-ray in / x-ray out*
- *better statistics than chopper*
- *Time-of-flight techniques*



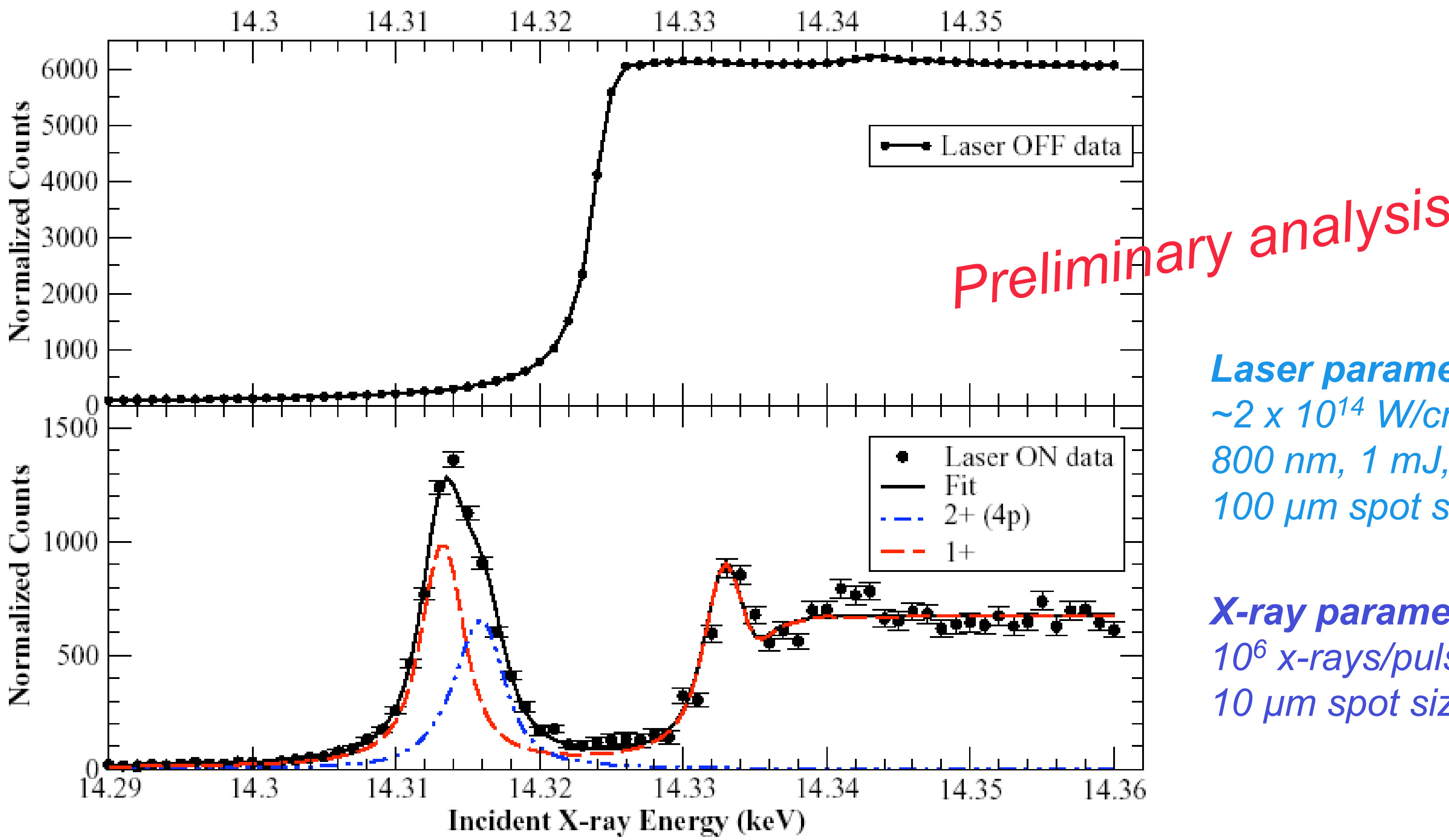
- *Compact,easy to use - no LN2*
- *Arrayable (8% of 4π w/one)*

Silicon Drift Detector



- *Hi count rate, hi resolution (~ 0.5 MHz, 240eV FWHM 8keV)*
- *Hi efficiency (300 μm thick) (70% @ 12 keV)*

Strong-field induced changes in Kr absorption

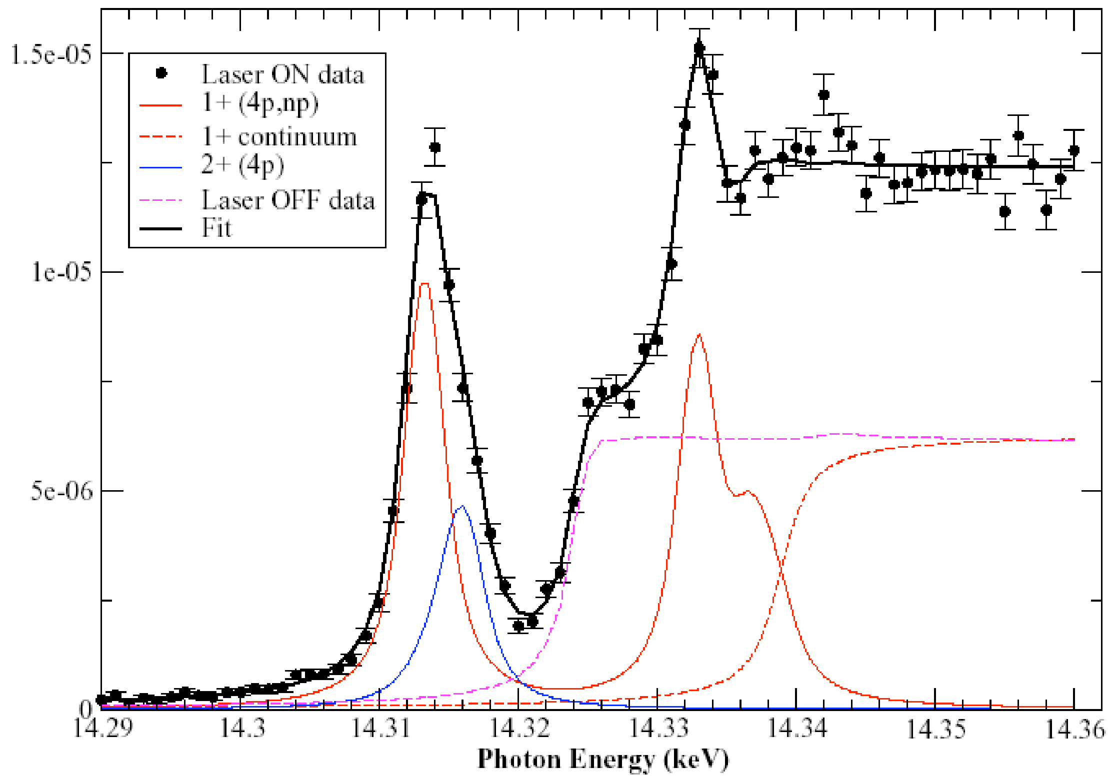


Laser parameters:
~2 x 10^{14} W/cm^2
800 nm, 1 mJ, 50 fs,
100 µm spot size

X-ray parameters:
10^6 x-rays/pulse
10 µm spot size

Extended scan range (7x) enabled by Dec 04 active stabilization of x-ray beam position to 1 µm during monochromator scan.

Strong-field modified Kr absorption spectrum



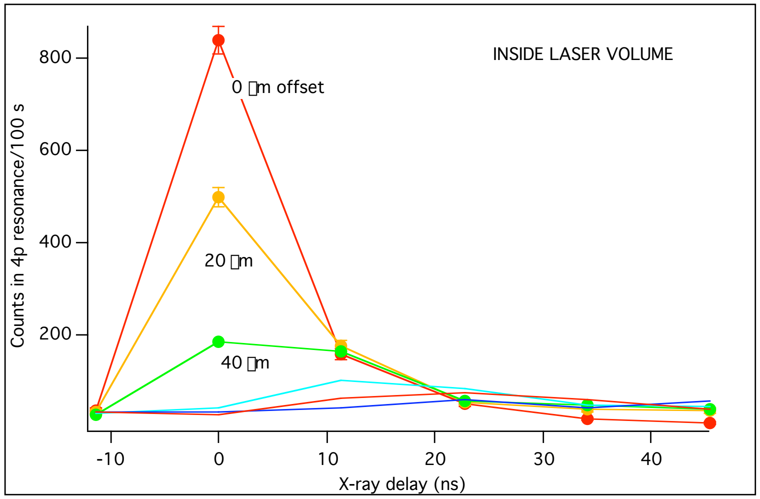
Kr+
122 transitions total
2 transitions to 4p
30 transitions to 5p
30 also to 6p, 7p, 8p

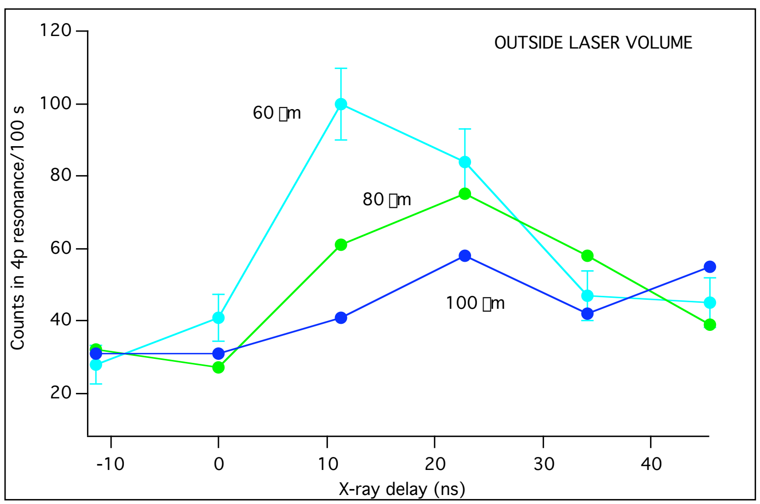
Kr2+
530 transitions total
14 transitions to 4p
129 transitions to 5p
129 also to 6p,7p,8p

Multiple open shells: [1s] [$4p^2$] 5p final state
Relativity and correlation simultaneously important

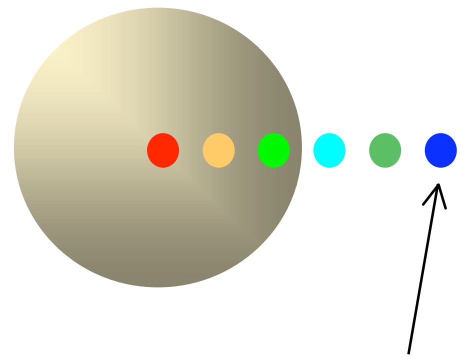
Theory: Lin Pan & Don Beck

Monitoring Coulomb Explosions with X-rays





Laser focus ~100 µm FWHM



X-ray probes
~10 µm

Average speed ~ 5 µm/ns
11 eV Kr+

Summary and outlook

- ***Goal: Understand changes to x-ray processes in presence of strong-laser fields***

- ***First step realized - x-ray microprobes of strong-field processes***
  - *Complete near-edge scan of Kr+*
  - *Coulomb expansion of "dense" ion cloud $10^{13}/cm^3$*
    - *energetic, anisotropic ion production*
    - *charge state decomposition & MD simulation ongoing*
  - *Dressed-atom ponderomotive shift experiment awaits*

    at 100 ps x-ray pulse length require 3μm focus for $10^{14} W/cm^2$

    at 1 ps x-ray pulse length require 30μm focus

- ***Short x-ray pulses on the horizon (1ps)***
  - *field-free molecular alignment studies (w/U Michigan)*
  - *coherent-control of molecular processes …*

Aligned molecules

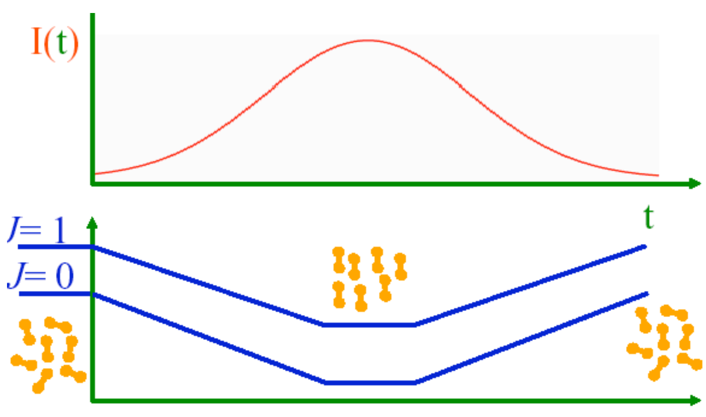
w/ S.T. Pratt, P.H. Bucksbaum

Motivations

- *Stringent tests of photoionization and photodissociation*
- *Study behavior of molecules as fcn of strength of aligning potential*
- *Aligned molecules aid single biomolecule structure determination (Hajdu et al)*
- *Test coherent control techniques to optimize alignment*

Adiabatic alignment

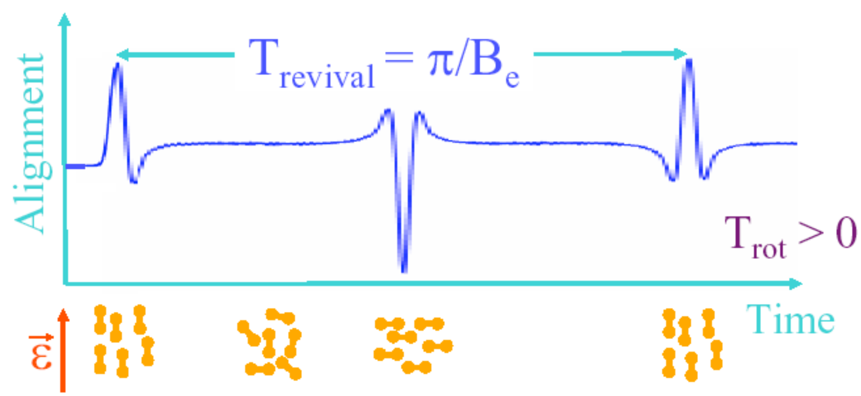
$\tau_{laser} > \tau_{rot}$



Friedrich & Hershbach, PRL 74, 4623 (95)

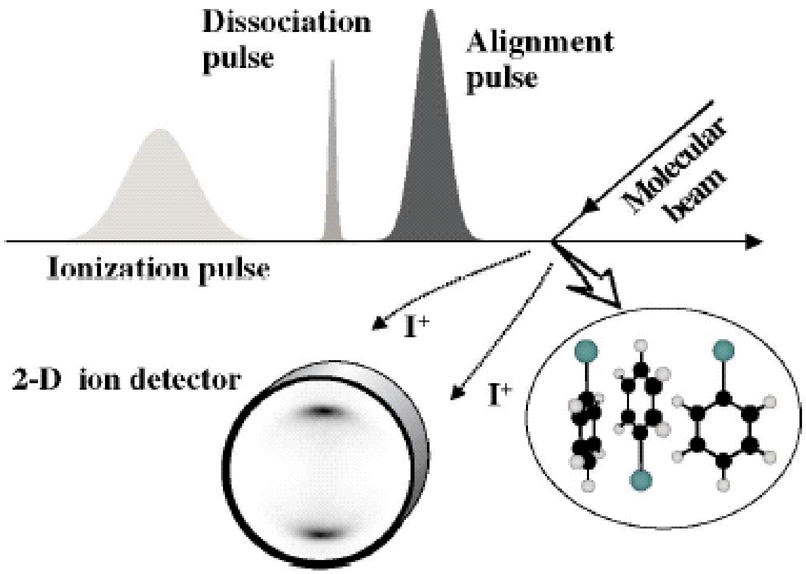
Field-free alignment

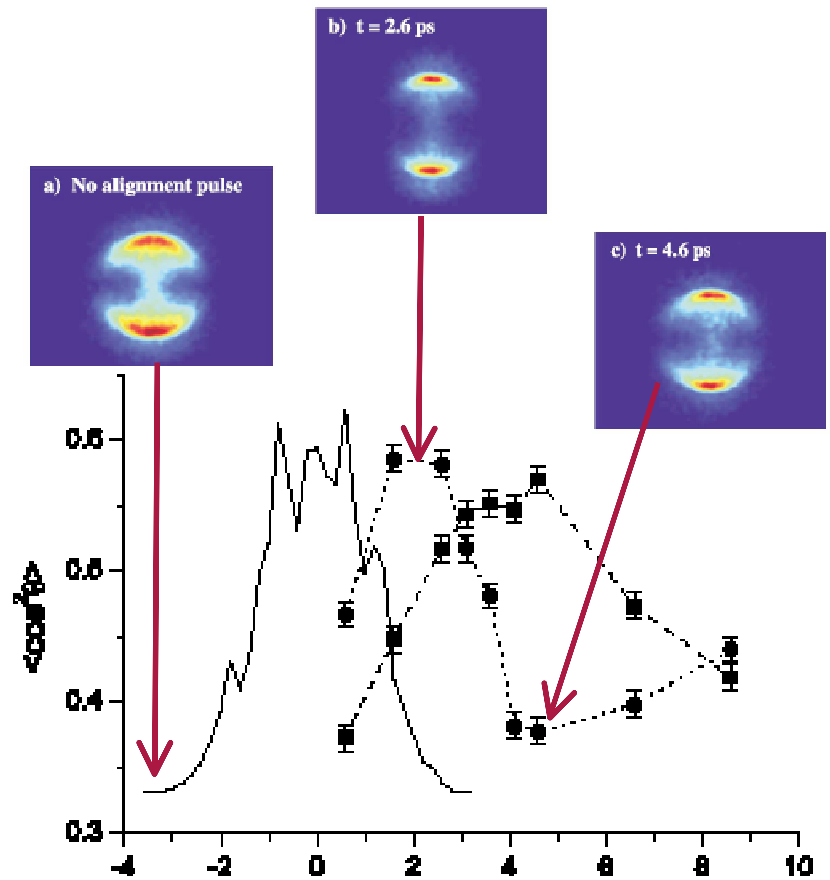
$\tau_{laser} < \tau_{rot}$



T. Seideman, PRL 83, 4971 (99)

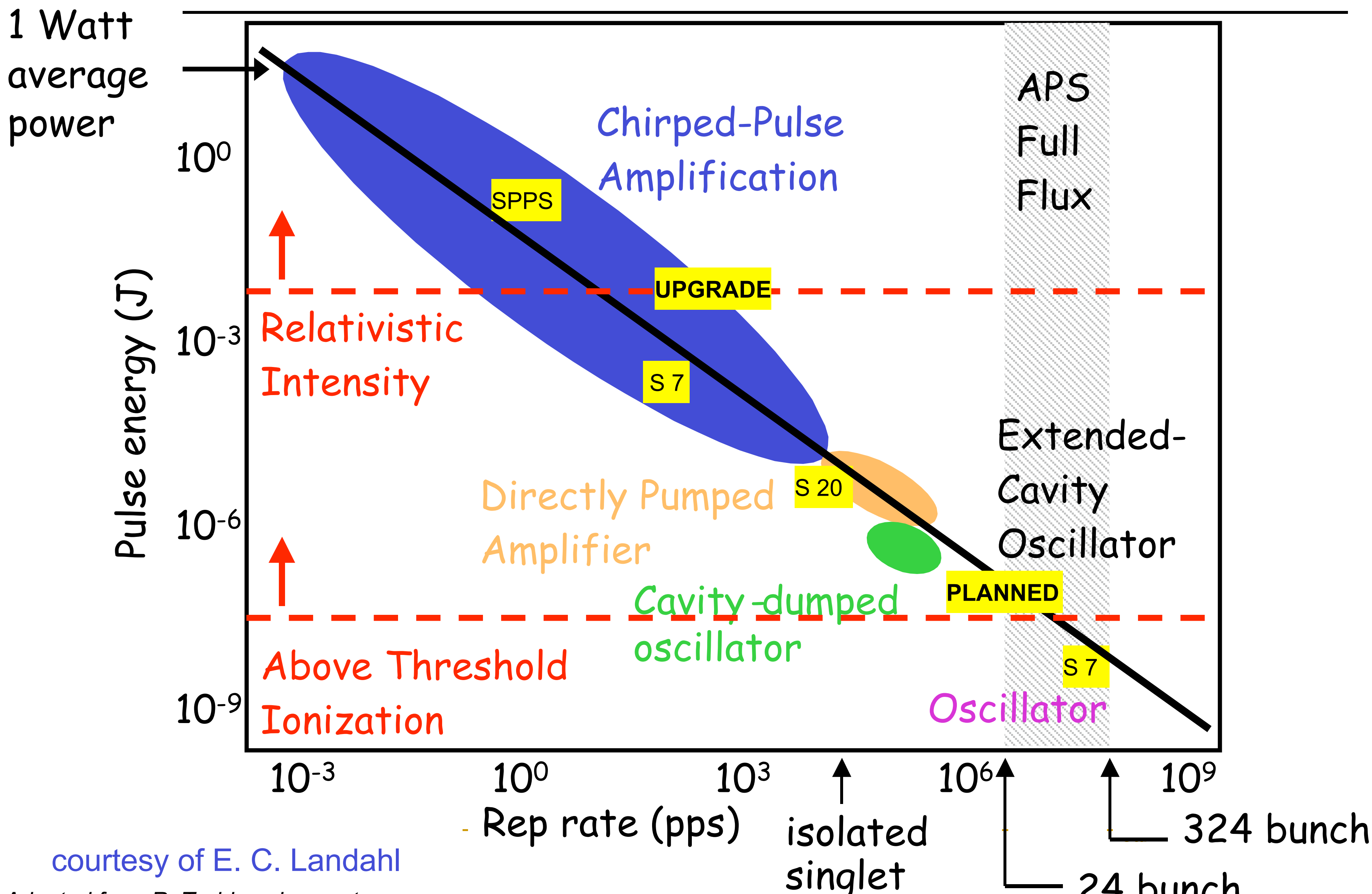
Field-free alignment studies need ps x-rays





Peronne et al.
Phys. Rev. Lett. **91**, 043003 (2003)

Femtosecond-laser pulse energy vs. repetition rate

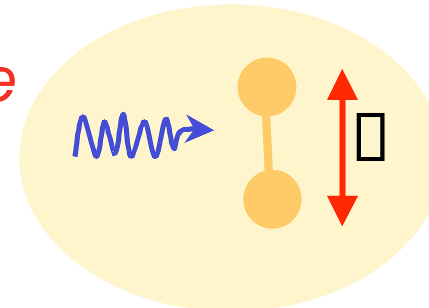


courtesy of E. C. Landahl

Adapted from R. Trebino class notes

Scientific wishlist - ultrafast AMO studies

Understand strong-field perturbed atomic structure
Field-free molecular alignment
Coherently-control molecular processes



Facility enablers

shorter pulses (~1 ps) - *dressed-atom studies w/current focus*
- *field-free molecular alignment*
- *coherent-control*

chopper for 24-bunch mode - *expand available beamtime*
soft x-ray energy range - *clearer atomic signatures*
polarization control - *dichroism studies*

Time-resolved instrumentation enablers

dispersive spectrometer + position sensitive detector
jitter-free streak camera
higher rep rate, high-power laser